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Uptake, Accumulation and Distribution of Potentially Toxic Trace Elements in Medicinal and Aromatic Plants

Dragoja Radanović[1*] • Svetlana Antić-Mladenović[2]

[1] Institute for Medicinal Plant Research "Dr Josif Pančić", Tadeuša Košćuška 1, 11000 Belgrade, Serbia
[2] University of Belgrade, Faculty of Agriculture, Nemanjina 6, 11080 Zemun, Serbia

Corresponding author: * **dradanovic@mocbilja.rs**

ABSTRACT

This paper reviews contents and distribution of essential and potentially toxic trace elements (Fe, Zn, Cu, Mn, Co, Cd, Pb, Cr, Ni) in medicinal and aromatic plants (MAPs). A large difference in metal uptake between plant species has been demonstrated in many studies. Concentration of trace elements in the MAPs is largely determined by soil type, growing site conditions and soil chemical processes, particularly soil pH. Additionally, anthropogenic pollution of soil and air affects metal contents in the MAPs. Furthermore, the species and genotypes of a species differ greatly in their ability for metal uptake. Recent studies have shown capability of some of the MAPs for metal hyperaccumulation; particularly *Hypericum perforatum* accumulates more Cd than other plants under the same growing conditions. Uptake, accumulation and translocation of metals, and consequently, a good quality of MAPs and final products that should be free from potentially harmful constituents, can be controlled by combining: (i) adequate choice of growing site (especially regarding: soil pH, organic matter content, clay content, redox potential, distance from point pollution sources, etc.), (ii) choice of suitable genotypes and (iii) appropriate management of soil and crops. Additionally, the MAPs, and in particular the plant parts to be utilized (e.g. roots, leaves, flowers, herb) should be regularly tested for contaminant load before processing for pharmaceutical drugs. The present review also highlights the potential risks of human intoxication with trace elements after the consumption of pharmaceutical drugs (essential oils, extracts, and teas).

Keywords: content, essential oil, heavy metals, herb, quality, pharmaceutical drug, soil characteristics, toxicity

CONTENTS

INTRODUCTION

Interest in medicinal and aromatic plants (MAPs) is increasing all over the world because of their lesser side effects as compared to the synthetic drugs besides cost effectiveness and easy availability. Thus, the quality of a certain species can be primarily estimated by its active component(s) that produce a desirable therapeutic response. Promoters of natural products have the opinion that herbal medicines are safe enough and even if the desirable therapeutic response is not achieved, their use is not dangerous to health because of their natural origin. However, the findings on the poisoning of trace elements - from different parts of the world have ruled out this principle of safety (Ernst 2002; Denholm 2010). World Health Organization (WHO) recommends that tests for toxic metals should be included in the specification for herbal materials (WHO guidelines on good manufacturing practice for herbal medicines 2007).

Trace elements are normal constituents in the terrestrial environment (Adriano 2001), thus they are found in all living organisms. Other terms that have been used and considered synonymous for trace elements are: trace metals, heavy metals (HMs), microelements and minor elements. Based on their role in living organisms, trace elements are divided in two groups: essential and non-essential elements. Essential elements (e.g. Fe, Mn, Cu, Zn) are required in trace amounts for the growth and development of living organisms and their deficiency might leads to metabolic malfunctioning. Non-essential elements (e.g. Hg, Pb, Cd) may be found in living organisms, however they are not required for regular biological functions. Rather, they adversely affect physiology and metabolism of all the living organisms (Kabata-Pendias and Mukherjee 2007).

Plants are considered as a key chain in the transfer of

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trace metals from soil to human beings. They absorb HMs from soil via roots, but under certain conditions aerial plant parts may be also involved. Uptake of metals usually depends on soil characteristics such as pH, organic matter content, cation exchange capacity, redox potential, salinity, as well as on the characteristics of specific plant species and their age. Beside during growth and development period, the MAPs are easily contaminated with HMs during harvesting operations and during drying, storage and processing of the produce.

In the global market, the MAPs mostly originate from spontaneous vegetation. The habitats of spontaneous MAPs are often located in non-cultivated and non-environment friendly areas, such as acid soils, alkaline soils and/or soils developed over ore deposits or metal-carrier minerals. The MAPs are cultivated on a rather small scale in various regions. Consequently, growing/collecting sites significantly differ in soil chemical and physical properties that affect metal mobility and availability in soils, and accordingly, the metal content in plants.

Soil reaction (pH) is one of the most responsible factors moderating solubility, mobility and availability of HMs in the soil (Adriano 2001). Mobility and availability of metals are generally low under neutral to basic soil reaction (Yoon *et al*. 2006). In soils with high pH and/or high content of lime (Ca-carbonates), availability of most metals decreases to such extent that, in a case of essential microelemets, it may lead to acute deficiency symptoms and retard in development of certain susceptible plants (e.g. Fe-chlorosis in *Arnica montana* and *Mentha arvensis* L.) (Subrahmanyam *et al.* 1992; Dachler and Pelzman 1999). In contrast, in acid conditions, the availability of most metals increases up to levels that cause their high accumulation. An example is high accumulation of Cd in various medicinal plants, especially St. John's wort (*Hypericum perforatum*) (Radanović *et al.* 2002; Chizzola and Lukas 2005) at low soil pH. There is also evidence of intensified Ni uptake by peppermint (*Mentha piperita*) grown in acid soil conditions (Radanović *et al.* 2001). In addition to pH, organic matter has paramount importance for decreasing or increasing metal mobility in soil (Mc Bride 1989).

Beside metal background level due to specific geo-climatic conditions, many soils have been additionally loaded with HMs from anthropogenic sources such as industrial activities, automobile exhaust, heavy-duty electric power generators, municipal wastes, refuze burning and use of pesticides in agriculture (Kabata-Pendias and Mukherjee 2007). The highest values reported regarding the most toxic metals, Cd and Pb, in the above ground parts (flowers, leaves and herb) of the MAPs are frequently associated with aero contamination and depositions (Zheljazkov and Nielsen 1996a, 1996b; Radanović *et al.* 2001; Barthwal *et al.* 2008; Zheljazkov *et al.* 2008a).

Furthermore, the MAPs represent a multitude of species with differing genotypic abilities for metal uptake. Also, plants can affect the metal mobility in soils by oxidizing their rhizosphere, excreting exudates and stimulating the activity of microbial symbionts in the rhizosphere, and consequently alter the available metal pool in a soil (Hinsinger 2001). Thus, big differences in the uptake of essential micronutrients and HMs are observed between the species and even within the varieties of the same species. Recent studies have shown that some MAPs, particularly St. John's wort, may show considerably higher content of Cd than other plants grown under the same conditions (Schneider *et al.* 2002). Based on the ability to accumulate Cd in the shoots, without detection of any toxicity symptoms, St. John's wort and some other MAPs (*Betula* sp., *Datura stramonium*, *Salicis* sp.) are considered as metal hyperaccumulators. Germany was among the pioneer countries that proposed reference values for HMs in the MAPs, above which their consumption may pose a threat to human health. The reference values for Cd and Pb are 0.2 and 5 mg kg^{-1} by DW, respectively (Schilcher 1994). For Hg, the reference value is 0.1 mg kg^{-1}. Cadmium reference values are comparatively higher for the species which take up the Cd preferentially. Accordingly, the reference value of Cd for the seeds of poppy (*Papaver* sp.), sunflower (*Helianthus annuus*) and linseed (*Linum usitatissimum* L.) is 0.8, 0.6 and 0.3 mg kg^{-1}, respectively (Chizzola *et al*. 2003). Similarly, the reference value of Cd for the herb of yarrow (*Achillea millefolium*) and St. John's wort is 0.3 and 0.5 mg kg^{-1}, respectively (Schilcher 1994).

An important aspect of the MAPs research is the HMs distribution between plant organs that have pharmaceutical and therapeutical values, such as root, shoot, leaves, flowers, berries and seeds. HMs, such as Pb, Cu and Cr, are generally poorly mobile in plants and, therefore, their content in roots is higher than in the shoots; however, some of them, such as Ni, Cd and Zn, are highly mobile within the plants and might easily relocate from the roots to the above-ground plant parts (Kabata-Pendias and Mukherjee 2007), thus posing a risk of building up in the components of the environment, which may ultimately lead to the contamination of the food chain. For example, Cd can reach concentrations as high as 1.4 and 1.5 mg kg^{-1} in the flowers of chamomile (*Chamomila recutita*) and seeds of *Borago officinalis*, respectively (Bingel *et al.* 1999). In addition, the harvest time may influence the heavy metal concentration in the herb. For example, comparatively a higher concentration of Cd was found in leaves and roots of buckwheat (*Fagopyrum esculentum*) at the second harvest than at the first harvest (Bingel *et al.* 1999).

Extractability of metals and their transfer from raw material to the final product to be consumed (e.g. herb teas, essential oils, etc.) is yet another concern related to the quality of the MAPs. Available data indicate significant differences in extraction efficiency between common extractants such as hot water, alcohol or propylene glycol. Among them, hot water has been identified as the least effective metal extractant (Radanović *et al.* 2007; Kostić *et al.* 2011). Distilled essential oils and herb teas are usually found to be free or low in heavy metal contents. Zheljazkov *et al.* (2008a) suggested that horehound (*Marrubium vulgare* L.), lemon balm (*Melissa officinalis* L.), and Greek oregano (*Origanum heracleoticum* L.) could be grown even on the heavy metal contaminated soils without any contamination of the marketable product, the essential oil.

Content of HMs in the MAPs and possible consequences are very complex. There should be awareness that HMs may be deliberately or accidentally included in the MAPs and herbal preparations. Furthermore, metals in the MAPs may be present in the concentrations that are beneficial to the users to correct micronutrient deficiencies or they may be present in concentrations that may pose threat to the consumer's health. In this review, we have discussed the plant contents of essential and potentially toxic trace elements (Fe, Zn, Cu, Mn, Co, Cd, Pb, Cr and Ni) in the MAPs as per records collected from different countries and environments. We have also discussed the factors affecting the uptake and accumulation of metals in the MAPs and the distribution of metals in various plant organs. Furthermore, risks of MAPs pollution from geogenic (natural) and anthropogenic sources are presented. Special emphasis has been given to Cd content in the MAPs and to the factors affecting Cd uptake. We have also covered the quality aspect of final product (essential oil, active constituents, etc.) obtained from the MAPs and the potential risk for human intoxication with HMs after consumption of the medicinal product.

ESSENTIAL MICRONUTRIENTS

Iron (Fe)

Common range of Fe content in different soils is between 0.1 and 10%. Iron distribution in soil profile is variable and controlled by several soil parameters. Soil texture, as well as soil minerals are the most determining factors of total soil-Fe. Iron constitutes about 5% by weight of the earth's crust and 3-4% of the soil, however, concentration of soluble Fe in a soil is extremely low (Schwertmann 1991). Iron solubility and availability in soils is strongly controlled by soil pH and redox status (Lindsay 1991). Excess of calcium carbonate ($CaCO_3$) in soil limits the Fe availability to such extent that may lead to acute deficiency. Main deficiency symptom manifested in plants is chlorosis of young plant organs. This phenomenon is known as "Fe-stress" (Lindsay and Schwab 1982). It was reported that mountain arnica (*Arnica montana*) might suffer from Fe-chlorosos if soil carbonate content exceeds 1.5%; therefore, only possible growth of this species is in acid soils (Dachler and Pelzman 1999). Japanese mint (*Mentha arvensis* L.) often suffers from Fe-chlorosis during the regeneration phase after first harvest when grown in alkaline/calcareous soils (Subrahmanyam *et al.* 1992).

The optimal content of Fe in plants is essential with respect to plant metabolism as well as nutrient supply to human beings and animals. The plant's ability to adsorb Fe varies with soil conditions, plant growth stages and properties of specific genotype. Yeritsyan and Economakis (2002) found enhanced uptake of Fe in oregano (*Origanum vulgare* spp. hirtum) with increasing Fe concentration in nutrient solution. The maximal uptake rate was recorded during the seed formation stage. Iron accumulation was higher in roots (1500-6740 mg kg^{-1} Fe) than in leaves (95-134 mg kg^{-1} Fe), while younger leaves contained comparatively the lower content of Fe than the older leaves.

The range of mean Fe content in the below ground plant parts (roots, bulbs, tubers, etc.) in selected food plants has been reported to vary from 34 – 46 and 3.3 – 3.7 mg kg^{-1} on fresh and dry weight basis, respectively (Kabata-Pendias and Mukherjee 2007). Green above-ground parts of plants (e.g. especially spinach) contain higher amounts of Fe than the other parts. Iron content in aboveground plant parts of different crops ranges from 30 to 100 mg kg^{-1}. Some plants like *Alyssum bertoloni* have high capability to adsorb Fe. They can accumulate up to 4000 mg kg^{-1} in the root and 1300 mg kg^{-1} in the leaves (Brooks 1998).

Some published information on Fe content in the MAPs from spontaneous habitats and/or from unpolluted soils is presented in **Table 1**. The differences in Fe content between

Table 1 Iron concentration (mg kg^{-1}) in various medicinal and aromatic plants from different countries.

MAP species	Plant part	Range[a]	Mean (± SD)[b]	Source/Country
Lavandula angustifolia	Flower	400 – 800[f]		Zheljazkov and Nielsen 1996a/Bulgaria
Arnica sp.	Flower	480 – 499		Radanović *et al.* 2004/Serbia
Calendulla officinalis	Flower	82.2 – 350	224.3	Antić-Mladenović *et al.* 2000/Serbia
Rosa canina	Fruit[c]		84 ± 1.9	Sekeroglu *et al.* 2008/Turkey
Vaccinium myrtilus	Fruit[c]	52 – 327	130.3	Jovančević *et al.* 2009/Montenegro
Rubus ideaus	Fruit[c]	86 – 235	143	Jovančević *et al.* 2009/Montenegro
Foeniculum vulgare	Fuit	43.8 – 109	60.7	Chizzola *et al.* 2003/Austria
Fam. *Asteraceae*	Herb[d]	13.4 – 307		Arceusz *et al.* 2010/Poland
Fam. *Apiaceae*	Herb	13.1 – 278		Arceusz *et al.* 2010/Poland
Fam. *Lamiaceae*	Herb	35.2 – 275		Arceusz *et al.* 2010/Poland
Mentha piperita	Herb	102 – 1154	305	Chizzola *et al.* 2003/Austria
Achilea millefolium	Herb	57.6 – 75.8	66.9	Chizzola *et al.* 2003/Austria
Hypericum perforatum	Herb	105 – 682	348	Radanović *et al.* 2004/Serbia
Hypericum perforatum	Herb	43 – 144.1	91.6	Antić-Mladenović *et al.* 2000/Serbia
Melissa officinalis	Herb	1565 – 4430	2997.5	Antić-Mladenović *et al.* 2000/Serbia
Melissa officinalis	Herb	126 – 1398	544	Chizzola *et al.* 2003/Austria
Ocimum basilicum	Herb	400 – 800[g]		Zheljazkov and Warman 2003/Canada
Ocimum basilicum	Herb		238.5 ± 0.033	Annan *et al.* 2010/Ghana
Tanacetum larvatum	Herb	2319 – 2321		Radanović *et al.* 2004/Serbia
Origanum onites	Herb		198 ± 2.4	Sekeroglu *et al.* 2008/Turkey
Euphorbia helioscopia L.	Herb		19.44	Hussain *et al.* 2011/Pakistan
Ranunculus mariculatus	Herb		85.02	Hussain *et al.* 2011/Pakistan
Urtica dioica	Herb		12.96	Hussain *et al.* 2011/Pakistan
Achilea millefolium	Herb	70.7 – 352	149.9	Antić-Mladenović *et al*. 2000/Serbia
Mentha piperita	Leaves	385 – 1045		Radanović *et al.* 1998/Serbia
Levisticum officinalis	Leaves	78 – 403	248	Chizzola *et al.* 2003/Austria
Salvia officinalis	Leaves	152 – 889	635	Chizzola *et al.* 2003/Austria
Plantago lanceolata	Leaves		777	Radanović *et al.* 2004/Serbia
Rosmarinus officinalis	Leaves		173 ± 3.2	Sekeroglu *et al.* 2008/Turkey
Ocimum basilicum	Leaves		390 ± 13.6	Sekeroglu *et al.* 2008/Turkey
Orig. vulgare spp.*hirtum*	Leaves	95 – 134[h]		Yeritsyan and Economakis 2002/Greece
Alchornea cordifolia	Leaves		33.0 ± 0.002	Annan *et al.* 2010/Ghana
Rauwolfia vomitoria	Leaves		574.0 ± 0.003	Annan *et al.* 2010/Ghana
Salvia officinalis	Leaves[e1]	84 – 1832	429.5	Maksimović *et al.* 1999/Serbia
Salvia officinalis	Leaves[e2]	215 – 2030	830	Maksimović *et al.* 1999 Serbia
Althea officinalis	Root		1399	Radanović *et al.* 2004/Serbia
Gentiana lutea	Root	675 – 1542	1032	Radanović *et al.* 2004/Serbia
Glycyrrhiza glabra	Root		175 ± 0.5	Sekeroglu *et al.* 2008/Turkey
Linum usitatissimum	Seed	46.5 – 105	71.7	Antić-Mladenović *et al*. 2000/Serbia
Mentha piperita	Stem	69 – 163		Radanović *et al.* 1998/Serbia
Origanum vulgare	Whole plant		152 ± 3.04	Kostić *et al.* 2011/Serbia
Calendula officinalis L.	Whole plant		137.53 ± 2.75	Kostić *et al.* 2011/Serbia
Saturea montana L.	Whole plant		264.24 ± 5.30	Kostić *et al.* 2011/Serbia

[a] – range as given in related source
[b] - mean (± standard deviation (SD)) as given in related source
[c] – on dry weight
[d]- the term "herb" refers to dry above-ground part of plants used for medicinal purposes
[e] - [e1] - native sage; [e2] – cultivated sage
[f]– lavender cultivated on uncontaminated and contaminated soils
[g] – in container experiment with high Cu compost application
[h] – in solution culture

and within the species are caused by conditions of growing sites and by genotype specifics.

Copper (Cu)

Average content of total Cu for different types of soils reported from all over the world ranges between 20 and 30 mg kg^{-1} (Alloway 1995). Contents of Cu are closely associated with soil texture, while soil pH and soil organic matter control its distribution and behavior. Thus, depending on soil properties, the range of average total Cu concentrations in soils may be more wide, from 8 mg kg^{-1} in acid sandy soils to 80 mg kg^{-1} in heavy loamy soils (Kabata-Pendias and Pendias 2001). Copper concentration in the soil solution is usually very low, ranging from 1×10^{-5} to 6×10^{-4} mol m^{-3} (Mengel and Kirkby 2001). Several soil variables control the solubility of Cu and its availability to plants; these include: pH, redox potential, organic matter, soil texture, mineral composition, temperature and water regime. The concentration of Cu in the soil solution decreases sharply with an increase in pH. Dissolved organic matter (DOM) has a great affinity to fix Cu. Solubility of Cu-organic complexes in the soil solution is less dependent on soil pH, therefore Cu uptake from alkaline soils occurs in the form of Cu-chelates (McBride 1989).

Copper contents in plants vary greatly and are controlled by several factors, of which Cu availability in soils and plant properties are the most significant ones. In most plant species, Cu concentration is low and is within the range 5-20 mg kg^{-1} DW (Mengel and Kirkby 2001). Average Cu content in various food plants show a relatively small variation. On fresh weight basis, it ranges from 0.1 to 3.2 mg kg^{-1} in vegetables, from 0.3 – 4 mg kg^{-1} in fruits, from 0.3 – 13 mg kg^{-1} in cereals and from 0.2 – 23.8 mg kg^{-1} in nuts (Ensminger *et al.* 1995). The deficiency of Cu strongly affects physiological processes. Chlorosis and tip necrosis of new leaves are considered to be well known Cu deficiency symptoms (Kabata-Pendias and Mukherjee 2007). On the other hand, Cu is considered quite toxic if absorbed by the plants in excess.

Copper concentration in MAPs also depends on the growing environment and plant species. Additionally, Cu is slightly mobile in plants; thus, higher Cu concentrations might be found in roots than in the above-ground parts. The range of Cu content in some of the MAPs is given in **Table 2**.

Concentration of Cu in the peppermint (*Mentha piperita* L.) grown on seven soil types in Serbia varied from 8 to 17

Table 2 Copper concentration (mg kg^{-1}) in various medicinal and aromatic plants (MAPs) reported from different countries.

MAP species	Plant part	Range[a]	Mean (±SD)[b]	Source/Country
Chamomila recutita	Flower	13.5 – 26.5[e]		Zheljazkov *et al.* 2008b/Bulgaria
Chamomila recutita	Flower		7 ± 0.1	Sekeroglu *et al.* 2008/Turkey
Lavandula angustifolia	Flower	20 – 40[f]		Zheljazkov and Nielsen 1996a/Bulgaria
Arnica sp.	Flower	10.2 – 11.9		Radanović *et al.* 2004/Serbia
Calendulla officinalis	Flower	15.2 – 23.5	18.3	Antić-Mladenović *et al.* 2000/Serbia
Coriandrum sativum	Fruit	11.5 – 26.2[e]		Zheljazkov *et al.* 2008b/Bulgaria
Rosa canina	Fruit[c]		24 ± 0.5	Sekeroglu *et al.* 2008/Turkey
Vaccinium myrtilus	Fruit[c]	6.3 – 12.9	9.1	Jovančević *et al.* 2009/Montenegro
Rubus ideaus	Fruit[c]	6.7 – 21.5	12.2	Jovančević *et al.* 2009/Montenegro
Foeniculum vulgare	Fuit	5.5 – 15.1	11.8	Chizzola *et al.* 2003/Austria
Mentha piperita	Herb[d]	6.3 – 10.3		Zheljazkov and Nielsen 1996b/Bulgaria
Mentha piperita	Herb	6.0 – 34.1		Chizzola *et al.* 2003/Austria
Achilea millefolium	Herb	15 – 27.3	20.1	Antić-Mladenović *et al.* 2000/Serbia
Achilea millefolium	Herb	4.9 – 10.3	6.8	Chizzola *et al.* 2003/Austria
Hypericum perforatum	Herb	9.0 – 18.6	14.6	Antić-Mladenović *et al.* 2000/Serbia
Hypericum perforatum	Herb	11 – 18		Radanović *et al.* 2002/Serbia
Melissa officinalis	Herb	5.0 – 13.3	8.4	Chizzola *et al.* 2003/Austria
Melissa officinalis	Herb	13.5 – 21.1[e]		Zheljazkov *et al.* 2008b/Bulgaria
Ocimum basilicum	Herb	22 – 46[g]		Zheljazkov and Warman 2003/Canada
Tanacetum larvatum	Herb	13.1 – 18.7		Radanović *et al.* 2004/Serbia
Origanum vulgare	Herb		7.3	Radanović *et al.* 2004/Serbia
Origanum onites	Herb		3 ± 1	Sekeroglu *et al.* 2008/Turkey
Euphorbia helioscopia L.	Herb		16.8	Hussain *et al.* 2011/Pakistan
Ranunculus mariculatus	Herb		11.0	Hussain *et al.* 2011/Pakistan
Urtica dioica	Herb		7.4	Hussain *et al.* 2011/Pakistan
Achilea millefolium	Herb	18 – 27		Radanović *et al.* 2002/Serbia
Mentha piperita	Leaves	8 – 17		Radanović *et al.* 1998/Serbia
Levisticum officinalis	Leaves	4.7 – 8.5	7.0	Chizzola *et al.* 2003/Austria
Salvia officinalis	Leaves	13.8 – 36.1[e]		Zheljazkov *et al.* 2008b/Bulgaria
Salvia officinalis	Leaves	5.7 – 17.5		Chizzola *et al.* 2003/Austria
Plantago lanceolata	Leaves		14	Radanović *et al.* 2004/Serbia
Ocimum basilicum	Leaves		28.0 ± 0.001	Annan *et al.* 2010/Ghana
Pegularia daemia	Leaves		114.5 ± 0.008	Annan *et al.* 2010/Ghana
Alchornea cordifolia	Leaves		11.5 ± 0.00	Annan *et al.* 2010/Ghana
Rauwolfia vomitoria	Leaves		9.35 ± 0.00	Annan *et al.* 2010/Ghana
Valeriana officinalis	Root		8.7	Radanović *et al.* 2004/Serbia
Gentiana lutea	Root	19 – 22	20.8	Radanović *et al.* 2007/Serbia
Glycyrrhiza glabra	Root		6 ± 0.4	Sekeroglu *et al.* 2008/Turkey
Linum usitatissimum	Seed	7.7 – 15	10.7	Antić-Mladenović *et al.* 2000/Serbia
Calendula officinalis L.	Whole plant		12.82 ± 0.26	Kostić *et al.* 2011/Serbia
Saturea montana L.	Whole plant		15.77 ± 0.31	Kostić *et al.* 2011/Serbia

[a] – range as given in related source
[b] - mean (± standard deviation (SD)) as given in related source
[c] – on dry weight
[d]- the term "herb" refers to dry above-ground part of plants used for medicinal purposes
[e] - at different distances from Pb – Zn smelter: 0.8; 3; and 9 km
[f]– lavender cultivated on uncontaminated and contaminated soils
[g] – in container experiment with high Cu compost application

mg kg^{-1}, depending on the soil type (Radanović *et al.* 1998), while a wider range of Cu (6 – 34.1 mg kg^{-1} Cu) has been reported in the peppermint from Austria regions (Chizzola *et al.* 2003).

Zheljazkov and Warman (2003) investigated the influence of relatively high Cu containing compost (over 1200 mg kg^{-1} Cu) on the growth and quality of basil (*Ocimum basilicum*). In comparison to the control, they reported enhanced yield of basil dry matter in all the compost treatments (20, 40, and 60% of soil volume); while, the Cu content in plant tissue was approximately at the same level (22, 26, 46, 28 mg kg^{-1} Cu). Copper in the essential oil of basil was below 0.25 mg L^{-1}.

Five species of medicinal plants (*Bidens tripartita* L., *Leonurus cardiaca* L., *Marrubium vulgare* L., *Melissa officinalis* L. and *Origanum heracleoticum* L.), when grown in Cu contaminated soils, revealed the following Cu distribution between the organs: roots > leaves > flower > stems (Zheljazkov *et al.* 2008a). Metal content in hot water extracts (teas) and infusions prepared from these species was relatively low (0.001 – 0.2 mg L^{-1}) and the essential oils were free of the metal.

Zinc (Zn)

Zinc concentration in uncontaminated soils ranges between 10 to 300 mg kg^{-1} with a mean of about 50 mg kg^{-1} (Malle 1992). Most of Zn is present in the lattice structure of primary and secondary minerals (Huang 1989). Zinc concentration in soil solution is very low, ranging from 3×10^{-5} to 5×10^{-3} mol m^{-3} (Barber 1984). Availability of Zn is highly dependent on soil pH and is very low at high soil pH. It is particularly low when $CaCO_3$ is present in a soil (Donner and Lynn 1989).

Table 3 Zinc concentration (mg kg^{-1}) in various medicinal and aromatic plants from different countries.

MAP species	Plant part	Range[a]	Mean (±SD)[b]	Source/Country
Chamomila recutita	Flower	31.1 – 197.0[e]		Zheljazkov *et al.* 2008b/Bulgaria
Chamomila recutita	Flower		21 ± 1.7	Sekeroglu *et al.* 2008/Turkey
Lavandula angustifolia	Flower	20 – 360[f]		Zheljazkov and Nielsen 1996a/Bulgaria
Arnica sp.	Flower	21.2 – 22.3		Radanović *et al.* 2004/Serbia
Calendulla officinalis	Flower	31.5 – 47.2	39.7	Antić-Mladenović *et al.* 2000/Serbia
Coriandrum sativum	Fruit	33.9 – 145[e]		Zheljazkov *et al.* 2008b/Bulgaria
Rosa canina	Fruit[c]		15 ± 0.5	Sekeroglu *et al.* 2008/Turkey
Vaccinium myrtilus	Fruit[c]	8.7 – 46.8	16.9	Jovančević *et al.* 2009/Montenegro
Rubus ideaus	Fruit[c]	13.1 – 58.7	24.7	Jovančević *et al.* 2009/Montenegro
Foeniculum vulgare	Fuit	19.5 – 63.5	37.4	Chizzola *et al.* 2003/Austria
Fam. *Asteraceae.*	Herb[d]	12.4 – 56.9		Arceusz *et al.* 2010/Poland
Fam. *Apiaceae*	Herb	18.1 – 53.0		Arceusz *et al.* 2010/Poland
Fam. *Lamiaceae*	Herb	13.3 – 85.3		Arceusz *et al.* 2010/Poland
Mentha piperita	Herb	14.4 – 42.4	28.7	Chizzola *et al.* 2003/Austria
Achilea millefolium	Herb	23 – 53		Radanović *et al.* 2002/Serbia
Achilea millefolium	Herb	23.6 – 53.2	33.9	Antić-Mladenović *et al.* 2000/Serbia
Achilea millefolium	Herb	20.5 – 40.5	28.5	Chizzola *et al.* 2003/Austria
Hypericum perforatum	Herb	21.2 – 59.3	42.7	Antić-Mladenović *et al.* 2000/Serbia
Hypericum perforatum	Herb	26 – 59		Radanović *et al.* 2002/Serbia
Melissa officinalis	Herb	13.5 – 66.1	32.2	Chizzola *et al.* 2003/Austria
Melissa officinalis	Herb	24.6 – 176.3[e]		Zheljazkov *et al.* 2008b/Bulgaria
Ocimum basilicum	Herb	32.6 – 191.8[e]		Zheljazkov *et al.* 2008b/Bulgaria
Origanum vulgare	Herb	15 – 21.1		Radanović *et al.* 2004/Serbia
Tanacetum larvatum	Herb	30 – 31.2		Radanović *et al.* 2004/Serbia
Origanum onites	Herb		9 ± 0.4	Sekeroglu *et al.* 2008/Turkey
Polygonatum verticillatum	Herb	38.8 – 60.0		Saeed *et al.* 2010/Pakistan
Euphorbia helioscopia L.	Herb		45.0	Hussain *et al.* 2011/Pakistan
Ranunculus mariculatus	Herb		28.6	Hussain *et al.* 2011/Pakistan
Urtica dioica	Herb		28.8	Hussain *et al.* 2011/Pakistan
Mentha piperita	Herb	38 – 197		Zheljazkov and Nielsen 1996b/Bulgaria
Mentha piperita	Leaves	20 – 50		Radanović *et al.* 1998/Serbia
Levisticum officinalis	Leaves	9.9 – 31.0	24.4	Chizzola *et al.* 2003/Austria
Salvia officinalis	Leaves	31.9 – 202.4[e]		Zheljazkov *et al.* 2008b/Bulgaria
Salvia officinalis	Leaves	21.6 – 44.9	33.0	Chizzola *et al.* 2003/Austria
Plantago lanceolata	Leaves		45.4	Radanović *et al.* 2004/Serbia
Taraxacum officinale	Leaves	25 – 70		Kranvogel *et al.* 1999/Germany
Rosmarinus officinalis	Leaves		9 ± 0.4	Sekeroglu *et al.* 2008/Turkey
Ocimum basilicum	Leaves		81.5 ± 0.001	Annan *et al.* 2010/Ghana
Desmodium adscendes	Leaves		164.5 ± 0.005	Annan *et al.* 2010/Ghana
Alchornea cordifolia	Leaves		46.5 ± 0.002	Annan *et al.* 2010/Ghana
Rauwolfia vomitoria	Leaves		47.5 ± 0.001	Annan *et al.* 2010/Ghana
Valeriana officinalis	Root		144.1	Radanović *et al.* 2004/Serbia
Taraxacum officinale	Root	12 – 35		Kranvogel *et al.* 1999/Germany
Gentiana lutea	Root		32.8 ± 1.3	Radanović *et al.* 2007/Serbia
Linum usitatissimum	Seed	40 – 63.2		Antić-Mladenović *et al.* 2000/Serbia
Cyperus rotunuds	Tuber	20 – 70		Haider *et al.* 2004/India
Calendula officinalis L.	Whole plant		18.15 ± 0.36	Kostić *et al.* 2011/Serbia
Saturea montana L.	Whole plant		25.12 ± 0.50	Kostić *et al.* 2011/Serbia

[a] – range as given in related source
[b] - mean (± standard deviation (SD)) as given in related source
[c] – on dry weight
[d]- the term "herb" refers to dry above-ground part of plants used for medicinal purposes
[e] - at different distances from Pb – Zn smelter: 0.8; 3; and 9 km
[f]– lavender cultivated on uncontaminated and contaminated soils

For most plant species, Zn concentrations in leaves below 10 to 15 mg kg^{-1} DW are indicative for Zn deficiency, whereas concentrations in the range of 20 to 100 mg kg^{-1} DW are considered sufficient (Mengel and Kirkby 2001). Worldwide, the mean Zn concentrations vary from 18 to 33 mg kg^{-1} by DW in cereal grains. In vegetables, Zn does not show excessive accumulation in vegetative plant parts, and its concentration is relatively low in fruits (Kabata-Pendias and Mukherjee 2007). Zinc deficiency in plants is observed at the contents less than 20 mg kg^{-1} and toxic effect might be expected when the concentration exceeds 300 – 400 mg kg^{-1}. The concentrations of Zn in the MAPs obtained from different sources are presented in **Table 3**.

Kranvogel *et al.* (1999) reported significant differences in Zn content in dandelion (*Taraxacum officinale*), depending on research design. They found average Zn content of 31.67 mg kg^{-1} in dandelions grown in natural habitats in Germany, while the plants grown in pots, containing the same soil, revealed 246.5 mg kg^{-1} of Zn (interval: 180 – 325 mg kg^{-1} Zn). Zinc concentration in spontaneous vegetation of dandelion was higher in the leaves (25 – 70 mg kg^{-1} Zn) compared to that in the roots (12 – 35 mg kg^{-1} Zn). High concentrations of Zn (approximately 70 mg kg^{-1} Zn) have been reported in the tubers of mustaka (*Cyperus rotundus*) from Bangalore and Punjab, India (Haider *et al.* 2004).

Pakade *et al.* (2011) measured Zn concentration in herbal drug mixtures, prepared from the plants collected from Hymalayan region. The lowest concentration of Zn (6.45 ± 1.55 mg kg^{-1}) was found in the mixture of *Curcuma longa*, *Zingiber officinale*, *Cochicum luteum*, *Withania somnifera*, Pine apple and *Boswellia serrata*, while the highest Zn concentration (22 ± 1.0 mg kg^{-1} Zn) was recorded in a mixture of *Withania somnifera*, *Centella asiatica*, *Gingko biloba*, *Bacopa monnieri*, *Ashatvarga*, *Pueraria tuberosa* and *Asparagus somnifera*.

Manganese (Mn)

Total Mn content may differ considerably in various soils, ranging from 10 to 9000 mg kg^{-1} (Kabata-Pendias and Pendias 2001). Globally, the mean Mn content in soils is estimated to range approximately from 437 to 600 mg kg^{1} (Kabata-Pendias and Mukherjee 2007). Manganese in soils occurs particularly in the ferromagnesian minerals. Its content in surface soils is positively associated with clay content. Soil pH and redox conditions are the master variables controlling the Mn dissolution in soils.

Concentrations of Mn in plants fluctuate greatly in res-

Table 4 Manganese concentration (mg kg^{-1}) in various medicinal and aromatic plants from different countries.

MAP species	Plant part	Range[a]	Mean (±SD)[b]	Source/Country
Chamomila recutita	Flower	258 – 293[e]		Zheljazkov *et al.* 2008b/Bulgaria
Chamomila recutita	Flower		21 ± 1.7	Sekeroglu *et al.* 2008/Turkey
Lavandula angustifolia	Flower	36 – 62[f]		Zheljazkov and Nielsen 1996a/Bulgaria
Arnica sp.	Flower	14.5 – 16.5		Radanović *et al.* 2004/Serbia
Calendulla officinalis	Flower	14 – 39	25	Antić-Mladenović *et al.* 2000/Serbia
Coriandrum sativum	Fruit	27.4 – 47.2[e]		Zheljazkov *et al.* 2008b/Bulgaria
Rosa canina	Fruit[c]		15 ± 0.5	Sekeroglu *et al.* 2008/Turkey
Vaccinium myrtilus	Fruit[c]	165 – 770	478	Jovančević *et al.* 2009/Montenegro
Rubus ideaus	Fruit[c]	130 – 848	350	Jovančević *et al.* 2009/Montenegro
Foeniculum vulgare	Fuit	23.8 – 45.0	32.6	Chizzola *et al.* 2003/Austria
Mentha piperita	Herb[d]	32.1 – 75.1	54.0	Chizzola *et al.* 2003/Austria
Achilea millefolium	Herb	25 – 172		Radanović *et al.* 2002/Serbia
Achilea millefolium	Herb	25.3 – 172	96.8	Antić-Mladenović *et al.* 2000/Serbia
Achilea millefolium	Herb	43.5 – 95.3	66	Chizzola *et al.* 2003/Austria
Hypericum perforatum	Herb	25 – 227	69.1	Antić-Mladenović *et al.* 2000/Serbia
Hypericum perforatum	Herb	25 – 226		Radanović *et al.* 2002/Serbia
Melissa officinalis	Herb	21.9 – 89.1	45.8	Chizzola *et al.* 2003/Austria
Melissa officinalis	Herb	159 – 321[e]		Zheljazkov *et al.* 2008b/Bulgaria
Ocimum basilicum	Herb	131 – 180[g]		Zheljazkov and Warman 2003/Canada
Tanacetum larvatum	Herb	55 – 109		Radanović *et al.* 2004/Serbia
Origanum vulgare	Herb	23.5 – 26.2		Radanović *et al.* 2004/Serbia
Origanum onites	Herb		9 ± 0.4	Sekeroglu *et al.* 2008/Turkey
Euphorbia helioscopia L.	Herb		70.0	Hussain *et al.* 2011/Pakistan
Ranunculus mariculatus	Herb		28.80	Hussain *et al.* 2011/Pakistan
Urtica dioica	Herb		40.80	Hussain *et al.* 2011/Pakistan
Mentha piperita	Herb	61.8 – 101		Zheljazkov and Nielsen 1996b/Bulgaria
Hyssopus officinalis	Inflorescence	33.4 – 65[e]		Zheljazkov *et al.* 2008/Bulgaria
Mentha piperita	Leaves	45 – 578		Radanović *et al.* 1998/Serbia
Levisticum officinalis	Leaves	48.2 – 124	78.9	Chizzola *et al.* 2003/Austria
Salvia officinalis	Leaves	291 – 348[e]		Zheljazkov *et al.* 2008b/Bulgaria
Salvia officinalis	Leaves	39 – 67.6	52.8	Chizzola *et al.* 2003/Austria
Plantago lanceolata	Leaves	82.6 – 86.5		Radanović *et al.* 2004/Serbia
Ocimum basilicum	Leaves		74.5 ± 0.001	Annan *et al.* 2010/Ghana
Alchornea cordifolia	Leaves		645 ± 0.05	Annan *et al.* 2010/Ghana
Rauwolfia vomitoria	Leaves		1455 ± 0.014	Annan *et al.* 2010/Ghana
Valeriana officinalis	Root	35 – 37		Radanović *et al.* 2004/Serbia
Gentiana lutea	Root	106 – 111	108.8 ± 1.9	Radanović *et al.* 2007/Serbia
Coix lachryma jobi	Root	50 – 200		Haider *et al.* 2004/India
Linum usitatissimum	Seed	21 – 38.2	30.7	Antić-Mladenović *et al.* 2000/Serbia
Calendula officinalis L.	Whole plant		24.38 ± 0.48	Kostić *et al.* 2011/Serbia
Saturea montana L.	Whole plant		40.65 ± 0.81	Kostić *et al.* 2011/Serbia

[a] – range as given in related source
[b] - mean (± standard deviation (SD)) as given in related source
[c] – on dry weight
[d]- the term "herb" refers to dry above-ground part of plants used for medicinal purposes
[e] - at different distances from Pb – Zn smelter: 0.8; 3; and 9 km
[f]– lavender cultivated on uncontaminated and contaminated soils
[g] – in container experiment with high Cu compost application

Table 5 Cobalt concentration (mg kg^{-1}) in various medicinal and aromatic plants from different countries.

MAP species	Plant part	Range[a]	Mean (±SD)[b]	Source/Country
Arnica sp.	Flower	2.0 – 2.1		Radanović *et al.* 2004/Serbia
Calendulla officinalis	Flower	0.5 – 2.5	2.0	Antić-Mladenović *et al.* 2000/Serbia
Vaccinium myrtilus	Fruit[c]	0.78 – 1.04	0.91	Jovančević *et al.* 2009/Montenegro
Rubus ideaus	Fruit[c]	0.95 – 2.03	1.42	Jovančević *et al.* 2009/Montenegro
Mentha piperita	Herb[d]	1.2 – 19		Radanović *et al.* 2004/Serbia
Hypericum perforatum	Herb	1 – 1.5	1.2	Antić-Mladenović *et al.* 2000/Serbia
Melissa officinalis	Herb	2.5 – 4.0	3.2	Antić-Mladenović *et al.* 2000/Serbia
Ocimum basilicum	Herb	0.9 – 1.2 [e]		Zheljazkov and Warman 2003/Canada
Tanacetum larvatum	Herb	3.5 – 5.0		Radanović *et al.* 2004/Serbia
Origanum vulgare	Herb	2.2 – 2.4		Radanović *et al.* 2004/Serbia
Achilea millefolium	Herb	1.5 – 2.0	1.7	Antić-Mladenović *et al.* 2000/Serbia
Salvia officinalis	Leaves	1.2 – 2.6	1.8	Maksimovic *et al.* 1999/Serbia
Plantago lanceolata	Leaves	2.4 – 2.6		Radanović *et al.* 2004/Serbia
Althea officinalis	Root	2.5 – 3.0		Radanović *et al.* 2004/Serbia
Gentiana lutea	Root	3 – 4	3.4 ± 0.5	Radanović *et al.* 2007/Serbia
Linum usitatissimum	Seed	1.0 – 1.5	1.3	Antić-Mladenović *et al.* 2000/Serbia

[a] – range as given in related source
[b] - mean (± standard deviation (SD)) as given in related source
[c] – on dry weight
[d]- the term "herb" refers to dry above-ground part of plants used for medicinal purposes
[e] - in container experiment with high Cu compost application

pect with plant type, plant organs and vegetative stage of plant growth (Bergman 1988; Mengel and Kirkby 2001). The average Mn concentration ranges from 27 to 50 mg kg^{-1} in cereal grains; while in vegetables, it ranges from 0.6 mg kg^{-1} (in tomatoes) to 20 mg kg^{-1} (in green peas) by FW (Kabata-Pendias and Mukherjee 2007). The critical Mn deficiency level, as recorded in the mature leaves of most plant species, ranges from 10 – 20 mg kg^{-1} by DW (Mengel and Kirkby 2001). Manganese toxicity, unlike deficiency, is not restricted to a narrow critical concentration range. Edwards and Asher (1982) reported values from 200 mg kg^{-1} in maize to 5300 mg kg^{-1} in sunflower that was associated with a 10% reduction of dry mater yield. Manganese toxicity is mainly reported in acid to very acid soils, where high concentrations of Mn^{2+}-ions prevail in soil along with flooding and a high reduction potential (Bergman 1988).

Manganese concentrations reported in the MAPs vary in a wide range (**Table 4**), depending on several factors, e.g. species, plant part, soil pH, etc. Smiechowska *et al.* (2004) found extremely high Mn concentration in various types of black tea leaves, with values ranging from 560 to 1366 mg kg^{-1}, as a result of high availability of Mn in very acid soils. Jovančević *et al.* (2009) found high amount of average Mn in dry matter of raspberry (350 mg kg^{-1} Mn) and blueberry (478 mg kg^{-1} Mn) fruits collected from different locations in Montenegro, with a very wide interval of variations (130 to 848 mg kg^{-1} Mn DW).

Radanović *et al.* (2001) found between 45 to 100 mg kg^{-1} Mn in peppermint plants, cultivated on seven different soil types in Serbia. An exceptionally high Mn content (578 mg kg^{-1}) was recorded in the peppermint plants grown in very acid Dystric Cambisol in a pot trial; whereas, the peppermint plants grown in the field trial showed much lower content of Mn (190 and 161 mg kg^{-1} DW, in leaves and stem, respectively). This investigation confirmed a significant negative correlation (r = 0.73**) between the soil pH and Mn content in peppermint.

Cobalt (Co)

Cobalt content in the soil is under strong influence of parent material and mechanical composition of the soil. Concentration of Co is usually higher in soils with high clay fraction, compared to the light sandy soils with significant amount of organic matter (Kabata-Pendias and Mukherjee 2007).

Cobalt is among the trace metals essential to plants. However, it is one of the most toxic elements if its concentration in a plant tissue exceeds a critical level. Depending on plant species and soil properties, critical Co concentration in plants varies from 15 to 30 mg kg^{-1} (Kabata-Pendias and Pendias 2001).

Although Co is considered essential for living organisms, information for MAPs is meager in scientific literature in comparison to other essential trace elements. Data reported from different countries in this regard (**Table 5**) show relatively small differences in Co concentrations between species (and plant parts).

TOXIC HEAVY METALS

Cadmium (Cd)

Cadmium is one of the most toxic metals to human beings. It is retained for many years in the human body. Therefore, consumption of foods high in Cd may induce chronic toxicity in human beings (Jackson and Alloway 1992). The global Cd content of soil has been estimated to range between 0.06 to 1.1 mg kg^{-1}, with an average of 0.5 mg kg^{-1} (Kabata-Pendias and Mukherjee 2007). In most soils, 99% of Cd is associated with soil colloids, thus very small proportion of Cd occurs in the soil solution (Christensen and Haung 1999). Anthropogenic sources of Cd in arable soils are of great concern at present. Considering all sources of Cd in soils, the major sources of pollution are atmospheric deposition and application of P-fertilizers.

Plants have no metabolic requirement for Cd, however its relatively easy availability to plants in Cd-rich soils poses serious health risk in human beings and livestock. Although several factors may control the uptake of Cd by plants, such as soil properties, weather, and plant species/cultivars, the soil Cd and soil pH are probably the most important ones (Eriksson *et al.* 1996). Cadmium content in the plants is mainly influenced by soil organic carbon and the soil pH; higher values of both soil organic carbon and pH tend to lower plant Cd content (Alloway 1995). Many authors have intensively studied cadmium content in the MAPs. Some published data in this regard are presented in **Table 6**.

According to recent studies, some of the MAPs, particularly St. John's wort, may show higher Cd content than other plants grown under the same conditions (Schneider *et al.* 2002). Since *Hypericum* species are characterized as Cd accumulators, there could be higher Cd concentrations in the plant than in the soil (Schneider and Marquard 1996). Several other MAPs, such as birch, buckwheat, dandelion, mallow, etc. have also been recognized as Cd accumulators (**Table 7**). Based on Cd availability in soils and Cd accumulation ability of plants, the earlier proposed guide-value for Cd content in some of the MAPs has risen from 0.2 to 0.3 mg kg^{-1} (*Crategus* sp., *Achilea millefolium*), and

Table 6 Cadmium concentration (mg kg^{-1}) in various medicinal and aromatic plants from different countries.

MAP species	Plant part	Range[a]	Mean (±SD)[b]	Source/Country
Tinospora cordifolia	as a raw material		9.8	Joseph *et al.* 2011/India
Zingiber officinale	as a raw material		8.8	Joseph *et al.* 2011/India
Chamomila recutita	Flower	0.27 – 11.9[e]		Zheljazkov *et al.* 2008b/Bulgaria
Chamomila recutita	Flower	0.4 – 1.45		Bingel *et al.* 1999/Germany
Chamomila recutita	Flower		0.19 ± 0.088	Radanović and Ant.-Mladenović 2008/Serbia
Lavandula angustifolia	Flower	0.1 – 13.3[f]		Zheljazkov and Nielsen 1996a/Bulgaria
Arnica sp.	Flower		0.20 ± 0.104	Radanović and Ant.-Mladenović 2008/Serbia
Calendulla officinalis	Flower	0.15 – 0.17		Radanović *et al.* 2000/Serbia
Tanacetum parthenium	Flower	0.10 – 0.45	0.28	Marković *et al.* 2008/Serbia
Coriandrum sativum	Fruit	0.23 – 8.9[e]		Zheljazkov *et al.* 2008b/Bulgaria
Anethum graveolens	Fruit		0.13 ± 0.031	Radanović and Ant.-Mladenović 2008/Serbia
Rosa canina	Fruit[c]		0.03 ± 0.003	Sekeroglu *et al.* 2008/Turkey
Vaccinium myrtilus	Fruit[c]	0.05 – 0.64	0.16	Antić-Mladenović *et al.* 2009/Serbia
Rubus ideaus	Fruit[c]	0.11 – 0.59	0.34	Antić-Mladenović *et al.* 2009/Serbia
Foeniculum vulgare	Fuit	<0.01 – 0.06	0.03	Chizzola *et al.* 2003/Austria
Mentha piperita	Herb[d]	0.02 – 0.27	0.05	Chizzola *et al.* 2003/Austria
Achilea millefolium	Herb	0.3 – 1.5		Radanović *et al.* 2002/Serbia
Achilea millefolium	Herb	0.09 – 0.39	0.21	Chizzola *et al.* 2003/Austria
Artemisia dracunculus	Herb		1.02	Marquard and Schneider 1995/Germany
Hypericum perforatum	Herb	0.15 – 0.98	0.59	Chizzola *et al.* 2003/Austria
Echinacea purpurea	Herb	0.05 – 0.1		Bingel *et al.* 1999/Germany
Hypericum perforatum	Herb	0.3 – 3.0		Radanović *et al.* 2002/Serbia
Hypericum perforatum	Herb	0.04 – 7.82		Schneider *et al.* 2002/Germany
Melissa officinalis	Herb	0.01 – 0.04	0.02	Chizzola *et al.* 2003/Austria
Melissa officinalis	Herb	0.26 – 10.2[e]		Zheljazkov *et al.* 2008b/Bulgaria
Ocimum basilicum	Herb		0.08 ± 0.004	Sekeroglu *et al.* 2008/Turkey
Ocimum basilicum	Herb	0.25 – 8.4[e]		Zheljazkov *et al.* 2008b/Bulgaria
Origanum vulgare	Herb		0.02 ± 0.007	Radanović and Ant.-Mladenović 2008/Serbia
Origanum onites	Herb		0.04 ± 0.001	Sekeroglu *et al.* 2008/Turkey
Viola tricolor	Herb		2.23	Marquard and Schneider 1995/Germany
Lippia multiflora	Herb		59.0 ± 0.007	Annan *et al.* 2010/Ghana
Euphorbia helioscopia L.	Herb		0.2	Hussain *et al.* 2011/Pakistan
Ranunculus mariculatus	Herb		0.6	Hussain *et al.* 2011/Pakistan
Chenopodium foliosum	Herb		0.4	Hussain *et al.* 2011/Pakistan
Mentha piperita	Herb	1.2 – 18.3[e]		Zheljazkov and Nielsen 1996b/Bulgaria
Mentha piperita	Leaves	0.05 – 0.44		Radanović *et al.* 2001/Serbia
Levisticum officinalis	Leaves	0.02 – 0.13	0.6	Chizzola *et al.* 2003/Austria
Salvia officinalis	Leaves	0.43 – 13.9[e]		Zheljazkov *et al.* 2008b/Bulgaria
Salvia officinalis	Leaves	< 0.1		Maksimović *et al.* 1999/Serbia
Salvia officinalis	Leaves	<0.01 – 0.03	0.01	Chizzola *et al.* 2003/Austria
Plantago lanceolata	Leaves		0.65 ± 0.358	Radanović and Ant.-Mladenović 2008/Serbia
Tanacetum parthenium	Leaves	0.25 – 0.55	0.41	Marković *et al.* 2008/Serbia
Ocimum basilicum	Leaves		32.0 ± 0.004	Annan *et al.* 2010/Ghana
Valeriana officinalis	Root		0.15 ± 0.071	Radanović and Ant.-Mladenović 2008/Serbia
Taraxacum officinale	Root	0.06 – 0.25		Kranvogel *et al.* 1999/Germany
Althea officinalis	Root		0.02 ± 0.001	Radanović and Ant.-Mladenović 2008/Serbia
Coix lachryma jobi	Root	0.43 – 1.71		Haider *et al*, 2004/India
Gentiana lutea	Root	0.5 – 0.75	0.7 ± 0.1	Radanović *et al.* 2007/Serbia
Calotropis procera	Root	0.1 – 0.25[g]		Barthwal *et al.* 2008/India
Linum usitatissimum	Seed	0.98 – 1.95		Bingel *et al.* 1999/Germany
Linum usitatissimum	Seed	0.24 – 0.26		Radanović *et al.* 2000/Serbia
Abutilon indicum	Seed	0.1 0.3[g]		Barthwal *et al.*2008/India
Cyperus rotundus	Tuber	0.32 – 1.40		Haider *et al.* 2004/India
Euphorbia hirta	Whole plant	0.12 –0.25[g]		Barthwal *et al.* 2008/India

[a] – range as given in related source
[b] - mean (± standard deviation (SD)) as given in related source
[c] – on dry weight
[d]- the term "herb" refers to dry above-ground part of plants used for medicinal purposes
[e] - at different distances from Pb – Zn smelter: 0.8; 3; and 9 km
[f]– lavender cultivated on uncontaminated and contaminated soils
[g] –plants grown at environmentally different sites

even up to 0.5 mg kg^{-1}, for Cd accumulating species (*Betula* sp., *H. perforatum*, *D. stramonium*, *Salicis* sp.). Out of 24 plants analyzed, Marquard and Schneider (1995) found tissue Cd content above the permissible level (0.2 mg kg^{-1}) in 11 medicinal plants in Germany. Moreover, in five species of medicinal plants (*H. perforatum*, *Hypericum hirsutum*, *Artemisia dracunculus* (G), *Artemisia dracunculus* (R) and *Viola tricolor*) they found Cd concentrations ranging from 1 to 2 mg kg^{-1}. Based on the ability of MAPs to accumulate HMs, Zheljazkov and Nielsen (1996b) and Zheljazkov and Warman (2003) considered the MAPs as potential phytoremediation option for the metal-polluted agricultural soils or as the recultivation option for degraded soils.

Research conducted on metal contaminated soils in Bulgaria (Zheljazkov *et al.* 2008a) distinguished bur-marigold (*Bidens tripartita* L.) as a potential plant species for high Cd accumulation (> 20 mg kg^{-1} of Cd in roots and >15 mg kg^{-1} of Cd in the leaves at contaminated sites; HNO_3 extractable soil Cd was 30 mg kg^{-1} at 0.5 km distance from the Pb-Zn smelter). Cadmium distribution within the plant has been reported to follow the order: roots > leaves > flowers > stems, with regard to several medicinal plants

Table 7 Cadmium content in selected Cd-accumulating medicinal plants (90% data from Kabelitz 1998, cited by Chizzola *et al.* 2003)

Plant (Latin name and common name)	Plant part	Cd (mg kg^{-1})
Chamomila recutita (Chamomile)	Flowers	0.42
Artemisia absinthium (Absinthe)	Herb	0.42
Solidago sp. (Goldenrod)	Herb	0.44
Nasturtium officinale (Watercress)	Herb	0.46
Achilea millefolium (Yarrow)	Herb	0.49
Linum usitatissimum (Linseed)	Seed	0.54
Taraxacum officinale (Dandelion)	Herb	0.69
Betula sp. (Birch)	Leaves	0.70
Fagopyrum esculentum (Buckwheat)	Herb	0.86
Malva sp. (Mallow)	Leaves	1.20
Hypericum perforatum (St. John's wort)	Herb	1.30
Salix sp. (Willow)	Bark	1.80

(*Bidens tripartita* L., *Leonurus cardiaca* L., *Marrubium vulgare* L., *Melissa officinalis* L. and *Origanum heracleoticum* L.). Similar order of distribution has been recorded regarding other HMs (Cu and Pb) within plants, though Cd shows greater mobility in plants in comparison to other metals. However, variable concentrations of Cd were found in hot water extracts (teas) and infusions (concentrated water extracts) prepared from the investigated species. Interestingly, the highest measured Cd concentration in tea and infusion was recorded in *Leonurus cardiaca*, and not in *B. triparttita*. Cadmium concentration in hot water extract prepared from different species was in the order: *L. cardiaca* > *B. tripartita* > *M. officinalis* > *Marubium vulgare* > *Origanum heracleoticum*. Zheljazkov *et al.* (2008a) concluded that selected medicinal crops might offer feasible agronomic, environmental and economic alternative to other commonly edible crops in areas where marketable products might be contaminated.

Research findings indicated that plants easily capture Cd from the soil, especially when growth medium contains high Cd levels. Much attention has been paid to soil properties, as well as to agrotechniques that affect Cd uptake. Many authors reported that soil pH, Cd content in a soil and soil organic carbon are crucial factors for Cd accumulation in the MAPs, especially in different *Hypericum* species (Plescher *et al.* 1995; Radanović *et al.* 2002; Chizzola and Lukas 2005). Radanović *et al.* (2002) found higher Cd contents in *Hypericum* species grown in acid soil than in the species grown in slightely acid to neutral soils. An inverse correlation between Cd uptake and pH of the soil (r= -0.72**) was found for *Mentha piperita*, as well (Radanović *et al.* 2001).

Transfer factor of Cd from soil to plant (the ratio of the metal content in plant tissue to the total concentration of metal in the soil) may vary greatly within the plant species and even in the variety or genotypes within a given species (Schneider *et al.* 2002). The varietal influence may depend on whether a given variety can retain Cd in the roots or translocate it to shoots.

Cadmium transfer from soil to plant may also be influenced by harvesting time. In various herb species, the higher Cd content was found after later harvesting than after earlier one (Schneider and Marquard 1996). Compared to the plants sown in autumn, the plants sown in spring yielded higher Cd content in the flower heads in *Chamomila recutita* (Pleschner *et al.* 1995). Chizzola *et al.* (2003) recommended careful choice of the growing site and the management of soil conditions in order to avoid enhanced Cd input into the food chain when producing poppy seeds and linseeds. There are also findings that fertilization with nitrogen and phosphorus might increase the Cd accumulation in St. John's wort (Azizi and Omidbaigi 2002).

As the concentration of Cd in MAPs reflects genetic differences among species/cultivars, and growing conditions affect Cd uptake and translocation, it is advisable to choose carefully the growing sites and to monitor Cd concentrations in plants at regular intervals. The latter is of special importance for minimizing herb contamination with Cd with regard to a field production of Cd hyperaccumulators, such as *Hypericum perforatum*.

Lead (Pb)

The natural Pb content in soils originates from parent material. Its abundance in sediments is a function of soil-clay content. Soils rich in clay fraction contain more Pb than the sandy soils. The overall average of total soil Pb for different soils is estimated to be 25 mg kg^{-1} (Kabata-Pendias and Pendias 2001). Unpolluted soils contain less than 100 mg kg^{-1} of Pb. Among various natural and anthropogenic sources of Pb contamination, the impact of industrial emission and use of leaded petrol are considered to be of the greatest environmental risk. When Pb is released into the environment from anthropogenic sources, it remains bioavailable for a long period, due to low solubility and relative low microbial degradation. The main soil pools for Pb are soil solution, adsorption surfaces of clay, humus exchange complex and secondary Fe/Mn oxides.

Lead is neither an essential nor a beneficial element for plants or animals, however, if present in excess, it can induce toxicity effects, resulting in stunted plant growth or even plant death. Plants can take up Pb both from soil and atmosphere. When plants are grown in unpolluted sites, Pb level is relatively stable. Lower contents of Pb (< 1 mg kg^{-1}) are recorded in cereal grains and fruits, while the higher ones (>1 mg kg^{-1} of Pb) are found in roots and tubes (Kabata-Pendias and Mukherjee 2007). Leafy vegetables and fodder plants usually contain higher Pb concentrations (>2 mg kg^{-1}). The scientific literature regarding soil factors influencing the uptake of Pb is often contradictory; however, there is a general view of low Pb availability in a soil. Lead mobility and availability in soils and its uptake by plants may be enhanced by the decrease of soil pH, organic matter, inorganic colloids, iron oxides and phosphorus content in a soil.

Lead concentration in the MAPs has been intensively investigated. Some reported results in this regard are presented in **Table 8**. Great variations in Pb content among different species have been reported in the scientific literature, the highest values in the above ground plant parts (flowers, leaves and herb) being mostly associate with aero-contamination and deposits (Zheljazkov and Nielsen 1996a, 1996b; Barthwal *et al.* 2008; Zheljazkov *et al.* 2008a). On the other hand, Pb content in the MAPs, grown at uncontaminated areas, mostly corresponds to its usual plant concentrations. Relative immobility of Pb in plants is also confirmed; in other words, Pb taken up from the soil tends to accumulate in plant roots. Root-biomass and leaf-chlorophyll content in some MAPs (e.g. *Matricaria chamomilla* L.) might be significantly reduced by Pb excess in growth medium (Grejtovský *et al.* 2008).

Zheljazkov and Nielsen (1996a) studied the effect of HMs (Cd, Pb, Cu, Mn, Zn, and Fe) originated from the anthropogenic source (Pb–Zn smelter in Plovdiv, Bulgaria) on the growth, productivity and quality of lavender (*Lavandula angustifilia* Mill.). The measured levels of Pb exceeded critical concentration in plant tissues (in all plant parts), yet without visible toxicity symptoms (Zheljazkov and Nielsen 1996a). The Pb concentrations in the plant parts were in the following order: stems > leaves = flowers > roots, ranging from 750 mg kg^{-1} in stems to 150 mg kg^{-1} in the roots. The main factor affecting the Pb distribution within the plant was aerocontamination at a distance of 500 m from the Pb-Zn smelter. It was estimated that 95% of Pb in the aboveground plant organs originate from the air. Additionally, Zheljazkov *et al.* (2008a) conducted a study on highly contaminated locations (>1000 mg kg^{-1} of total soil Pb), but at a certain distance from the direct aero-contamination source. They reported a relatively low Pb bioavailability factor (< 3%) and very low Pb transfer factor (0.01%) for the investigated MAPs (*Bidens tripartita* L., *Leonurus cardiaca* L., *Marrubium vulgare* L., *Melissa*

Table 8 Lead concentration (mg kg^{-1}) in various medicinal and aromatic plants from different countries.

MAP species	Plant part	Range[a]	Mean (±SD)[b]	Source/Country
Tinospora cordifolia	as a raw material		58.0	Joseph *et al.* 2011/India
Zingiber officinale	as a raw material		70.0	Joseph *et al.* 2011/India
Chamomila recutita	Flower	4.2 – 149.6[e]		Zheljazkov *et al.* 2008b/Bulgaria
Chamomila recutita	Flower		2.9 ± 1.237	Radanović and Ant.-Mladenović 2008/Serbia
Lavandula angustifolia	Flower	1 – 550[f]		Zheljazkov and Nielsen 1996a/Bulgaria
Arnica sp.	Flower		0.81 ± 0.227	Radanović and Ant.-Mladenović 2008/Serbia
Calendulla officinalis	Flower	2.5 – 12	4.7	Radanović *et al.* 2000/Serbia
Tanacetum parthenium	Flower	0.4 – 5.65	2.60	Marković *et al.* 2008/Serbia
Coriandrum sativum	Fruit	4.1 – 143.8[e]		Zheljazkov *et al.* 2008b/Bulgaria
Anethum graveolens	Fruit		3.25 ± 0.748	Radanović and Ant.-Mladenović 2008/Serbia
Vaccinium myrtilus	Fruit[c]	1.6 – 10.5	4.0	Antić-Mladenović *et al.* 2009/Serbia
Rubus ideaus	Fruit[c]	2.15 – 6.05	3.37	Antić-Mladenović *et al.* 2009/Serbia
Foeniculum vulgare	Fuit	0.1 – 1.1	0.4	Chizzola *et al.* 2003/Austria
Mentha piperita	Herb	0.2 – 2.4	0.8	Chizzola *et al.* 2003/Austria
Achilea millefolium	Herb[d]	2.5 – 8.0		Radanović *et al.* 2002/Serbia
Achilea millefolium	Herb	0.2 – 1.7	1.0	Chizzola *et al.* 2003/Austria
Hypericum perforatum	Herb	0.2 – 1.0	0.4	Chizzola *et al.* 2003/Austria
Hypericum perforatum	Herb	0.5 – 3.5		Radanović *et al.* 2002/Serbia
Melissa officinalis	Herb	0.5 – 1.6	0.8	Chizzola *et al.* 2003/Austria
Melissa officinalis	Herb	3.5 – 126.7[e]		Zheljazkov *et al.* 2008b/Bulgaria
Ocimum basilicum	Herb	4.5 – 525[e]		Zheljazkov *et al.* 2008b/Bulgaria
Origanum vulgare	Herb		0.63 ± 0.353	Radanović and Ant.-Mladenović 2008/Serbia
Euphorbia helioscopia L.	Herb		2.2	Hussain *et al.* 2011/Pakistan
Ranunculus mariculatus	Herb		1.2	Hussain *et al.* 2011/Pakistan
Urtica dioica	Herb		1.4	Hussain *et al.* 2011/Pakistan
Mentha piperita	Herb	7.4 – 173.2		Zheljazkov and Nielsen 1996b/Bulgaria
Datura stramonium	Leaves		0.89 ± 0.02	Olowoyo *et al.* 2012/South Africa
Amaranthus spinosus	Leaves		1.98 ± 0.08	Olowoyo *et al.* 2012/South Africa
Mentha piperita	Leaves	0.5 – 6.0		Radanović *et al.* 2001/Serbia
Levisticum officinalis	Leaves	0.4 – 4.3	1.9	Chizzola *et al.* 2003/Austria
Salvia officinalis	Leaves	1.2 – 34.4	7.40	Maksimović *et al.* 1999/Serbia
Salvia officinalis	Leaves	6.1 – 182.9[e]		Zheljazkov *et al.* 2008/Bulgaria
Salvia officinalis	Leaves	0.6 – 1.3	0.8	Chizzola *et al.* 2003/Austria
Plantago lanceolata	Leaves		1.12 ± 0.37	Radanović and Ant.-Mladenović 2008/Serbia
Cymbopogon citratus	Leaves	8.5 – 12.2		Haider *et al.* 2004/India
Tanacetum parthenium	Leaves	0.8 – 7.8	3.15	Marković *et al.* 2008/Serbia
Valeriana officinalis	Root		4.15 ± 1.474	Radanović and Ant.-Mladenović 2008/Serbia
Althea officinalis	Root		3.85 ± 1.328	Radanović and Ant.-Mladenović 2008/Serbia
Coix lachryma jobi	Root	9.3 – 13.9		Haider *et al.* 2004/India
Gentiana lutea	Root	4.5 – 5.5	5.1 ± 0.5	Radanović *et al.* 2007/Serbia
Calotropis procera	Root	0.8 – 2.2[g]		Barthwal *et al.* 2008/India
Datura stramonium	Root		2.13 ± 0.02	Olowoyo *et al.* 2012/South Africa
Amaranthus spinosus	Root		1.31 ± 0.02	Olowoyo *et al.* 2012/South Africa
Linum usitatissimum	Seed	0.5 – 1.5		Radanović *et al.* 2000/Serbia
Cardiospermum halicac	Seed	5.8 – 9.3		Haider *et al.* 2004/India
Abutilon indicum	Seed	0.6 – 1.15[g]		Barthwal *et al.* 2008/India
Cyperus rotundus	Tuber	14.9 – 18.5		Haider *et al.* 2004/India
Euphorbia hirta	Whole plant	1.0 – 1.9[g]		Barthwal *et al.* 2008/India

[a] – range as given in related source
[b] - mean (± standard deviation (SD)) as given in related source
[c] – on dry weight
[d]- the term "herb" refers to dry above-ground part of plants used for medicinal purposes
[e] - at different distances from Pb – Zn smelter: 0.8; 3; and 9 km
[f]– lavender cultivated on uncontaminated and contaminated soils
[g] –plants grown at environmentally different sites

officinalis L. and *Origanum heracleoticum* L.). They found that the roots were the main organ of Pb accumulation, while the lowest Pb concentrations were found in the flowers and steam (< 10 mg kg^{-1} of Pb). The Pb concentrations in essential oils were below detection limit; similarly, Pb in teas and infusions were very low, ranging from undetectable amounts to 0.02 mg L^{-1}.

Barthwal *et al.* (2008) reported a different pattern of Pb accumulation in *Euphorbia hirta* and *Peristrophe bycaliculata* exposed to the same environmental conditions in heavy traffic area. Haider *et al.* (2004) also reported different Pb levels in the MAPs collected from the same region. These findings imply that, besides the site specific factors, Pb uptake depends on genotype characteristics. Therefore, a generalized approach in estimation of heavy metal load in the MAPs should be avoided and each medicinal plant should be tested separately.

In quality investigations of herbal drugs, prepared from plants collected from Himalayan region, Pakade *et al.* (2011) found relatively similar Pb concentrations in two herbal mixtures (6 ± 0.66 mg kg^{-1} of Pb in the mixture of *Withania somnifera*, *Centella asiatica*, *Gingko biloba*, *Bacopa monnieri*, *Ashatvarga*, *Pueraria tuberosa* and *Asparagus somnifera*) and 4 ± 0.7 mg kg^{-1} of Pb in the mixture of *Cyperus rotundus*, *Coelus forskohlii*, *Rubia cardiofolia*, *Tinospora cordiofolia* Trifla and *Azadirachta indica*. According to Marković *et al.* (2008), Pb content in some MAPs might also be influenced by the sowing time (spring/autumn), even at the level of population within the same species.

Chromium (Cr)

Chromium is an essential micronutrient for human beings and animals. It controls the metabolism of glucose and lipids, but there is no clear evidence of Cr essentiality to plant metabolism. In contrast, the detrimental effects of Cr on plant growth and development are well documented (Shanker *et al.* 2005). Median content of Cr in various soils has been reported as 54 mg kg^{-1}. Its content in soils is determined mainly by parent material composition, with higher contents being generally observed in the soils derived from mafic and ultramafic rocks. A positive relationship between Cr and granulometric soil fractions has been reported; hence, a higher Cr content can be found in silty and loamy soils than in sandy soil (Kabata-Pendias and Pendias 2001). Additionally, numerous anthropogenic activities over the past decades have led to an increase in soil Cr above the natural (geogenic and background) levels at many sites, resulting in a widespread soil and environment pollution of Cr.

The most stable and common oxidation states of Cr in nature are Cr (III) and Cr (VI). These oxidation states have contrasting chemical properties and behavior, with respective environmental implications. Chromium (VI) exists as an anion. It is readily extracted from the soil and is considered to be much toxic form, while Cr (III) is relatively insoluble and less mobile. However, Cr (VI) is very unstable under normal soil conditions. It is a strongly oxidizing form, and in the presence of soil organic matter and/or Fe(II), it is rapidly reduced to Cr (III). Such a reduction is more rapid in acid than alkaline soils. Thus, in the majority of the soils, relatively less harmful form of Cr [Cr (III)] predominates. Oxidation of a proportion of Cr (III) is considered likely in soils with pH greater than 5 and rich in oxidized Mn (Alloway 1995). The form of Cr most available to plants is Cr (VI).

Chromium is not easily translocated within plants and is accumulated mainly in roots. Non-polluted or background Cr concentrations in plants are generally less than 1 mg kg^{-1} even across a wide range of soil-Cr values. The Cr content in cereal grains vary from 0.01 to 0.41 mg kg^{-1}, while the range for leafy vegetables and fruits is 0.04-0.08 mg kg^{-1} Cr (Kabata-Pendias and Mukherjee 2007).

Chromium content in the MAPs also vary in a wide range (**Table 9**), mostly depending on the characteristics of growing sites and plant genetic potential. Elevated content of Cr in *Millefoli herba*, and in the roots of *Valeriana officinalis*, *Angelica archangelica*, *Levisticum officinale*, *Gentiana lutea* and *Althea officinalis* has been reported for the areas under the influence of serpentinic minerals (Serbia), while the MAPs collected from non-contaminated calcareous Chernozem revealed usual range of Cr in plant tissue (Radanović *et al.* 2000). The effects of growing sites on Cr content in different MAPs have been discussed in detail by Haider *et al.* (2004) and Barthwal *et al.* (2008).

The influence of soil chemical properties, such as pH, redox potential, organic matter, etc., on Cr status has been intensively studied in agricultural plants, however, the information related to the MAPs are very poor. Radanović *et al.* (2001) found a positive correlation between soil-humus and Cr in peppermint leaves, which was probably associated with soil organic matter composition and formation of soluble and mobile, and thus plant-available organic

Table 9 Chromium concentration (mg kg^{-1}) in various medicinal and aromatic plants from different countries.

MAP species	Plant part	Range[a]	Mean (±SD)[b]	Source/Country
Tinospora cordifolia	as a raw material		53.8	Joseph *et al.* 2011/India
Zingiber officinale	as a raw material		318.8	Joseph *et al.* 2011/India
Chamomila recutita	Flower		0.75 ± 0.071	Radanović and Ant.-Mladenović 2008/Serbia
Arnica sp.	Flower		20.8 ± 11.65	Radanović and Ant.-Mladenović 2008/Serbia
Calendulla officinalis	Flower	0.5 – 3.5		Radanović *et al.* 2000/Serbia
Tanacetum parthenium	Flower	1 – 2.1	1.45	Marković *et al.* 2008/Serbia
Anethum graveolens	Fruit		1.0 ± 0.25	Radanović and Ant.-Mladenović 2008/Serbia
Vaccinium myrtilus	Fruit[c]	0.25 – 1.37	0.91	Antić-Mladenović *et al.* 2009/Serbia
Rubus ideaus	Fruit[c]	0.21 – 0.87	0.54	Antić-Mladenović *et al.* 2009/Serbia
Origanum vulgare	Herb[d]		16.9 ± 4.89	Radanović and Ant.-Mladenović 2008/Serbia
Euphorbia helioscopia L.	Herb		50.6	Hussain *et al.* 2011/Pakistan
Ranunculus mariculatus	Herb		24.2	Hussain *et al.* 2011/Pakistan
Urtica dioica	Herb		29.8	Hussain *et al.* 2011/Pakistan
Ocimum basilicum	Herb	10.3 – 11.0[e]		Zheljazkov and Warman 2003/Canada
Datura stramonium	Leaf		7.31 ± 0.12	Olowoyo *et al.* 2012/South Africa
Amaranthus spinosus	Leaf		6.74 ± 0.01	Olowoyo *et al.* 2012/South Africa
Mentha piperita	Leaves	1.75 – 6.3		Radanović *et al.* 2001/Serbia
Salvia officinalis	Leaves	1.0 – 9.0	2.9	Maksimović *et al.* 1999/Serbia
Plantago lanceolata	Leaves		11.8 ± 6.39	Radanović and Ant.-Mladenović 2008/Serbia
Cymbopogon citratus	Leaves	1.68 – 5.30		Haider *et al.* 2004/India
Tanacetum parthenium	Leaves	0.7 – 2.6	1.79	Marković *et al.* 2008/Serbia
Tanacetum parthenium	Leaves	0.8 – 7.8	3.15	Marković *et al.* 2008/Serbia
Valeriana officinalis	Root	6.8 – 32.2	19.5	Radanović and Ant.-Mladenović 2008/Serbia
Angelica arhangelica	Root		47.1 ± 23.5	Radanović and Ant.-Mladenović 2008/Serbia
Althea officinalis	Root	1.25 – 6.20	3.72	Radanović and Ant.-Mladenović 2008/Serbia
Coix lachryma jobi	Root	4.22 – 12.58		Haider *et al.* 2004/India
Gentiana lutea	Root	11.5 – 16.5	14.1 ± 2.1	Radanović *et al.* 2007/Serbia
Calotropis procera	Root	0.2 – 1.25[f]		Barthwal *et al.* 2008 India
Datura stramonium	Root		14.05 ± 0.89	Olowoyo *et al.* 2012/South Africa
Amaranthus spinosus	Root		18.31 ± 0.01	Olowoyo *et al.* 2012/South Africa
Linum usitatissimum	Seed	0.25 – 0.45		Radanović *et al.* 2000/Serbia
Cardiospermum halicac	Seed	3.19 – 9.64		Haider *et al.* 2004/India
Abutilon indicum	Seed	0.1 – 1.21[f]		Barthwal *et al.* 2008/India
Cyperus rotundus	Tuber	1.61 – 22.17		Haider *et al.* 2004/India
Euphorbia hirta	Whole plant	0.4 – 0.65[f]		Barthwal *et al.* 2008/India

[a] – range as given in related source
[b] - mean (± standard deviation (SD)) as given in related source
[c] – on dry weight
[d]- the term "herb" refers to dry above-ground part of plants used for medicinal purposes
[e] - in container experiment with high Cu compost application
[f]– plants grown at environmentally different sites

Cr-complexes at the investigated sites.

Similar to Pb, once in a plant, Cr has a tendency to accumulate in roots tissues. Root of some MAPs (e.g. *Valeriana* sp. and *Gentiana* sp.) is a raw material for further utilisation and/or production of pharmaceutical drugs. Consequently, high content of HMs in root tissues raises up a question of quality of pharmaceutical drugs, and of associated health risk. Radanović *et al.* (2007) studied the relationship between high Cr concentration (14 mg kg^{-1} of Cr by DW) in the root of gentian (*Gentiana lutea*) and the root-based galenic forms (ethanol extract, sppisum and siccum). They found low Cr-extrating efficiency of ethanol from the gentian roots; thus, Cr intake from such extracts might be considered negligible.

Nickel (Ni)

The average concentration of Ni in various soils of the world is around 20 mg kg^{-1}. However, Ni content in a particular soil is highly dependent on its content in parent material, apart from the soil-forming processes and pollution. Geochemically elevated Ni contents are observed in the soils derived from mafic and ultamafic rocks and especially in the soils derived from serpentine minerals, in which Ni content ranges from 100 to 7000 mg kg^{-1} (Brooks 1987). Amongst anthropogenic sources of Ni in soils, the largest one is burning of fuel and residual oils, followed by coal combustion and Ni mining and smelting. Some agricultural materials, such as sewage sludge, can also contribute to soil pollution with Ni (Alloway 1995).

Nickel in soils is slightly mobile. Several soil properties, particularly clay fraction, organic matter content and pH, control the behavior of Ni and its bioavailability in the soil. Generally, Ni mobility is inversely related to soil pH. Siebielec and Chaney (2006) reported a drastic increase of Ni extractability in the soil with pH < 6.5. In surface soils, Ni is bound to organic forms, a part of which is relatively soluble by chelates.

The content of Ni in plants is controlled by Ni origin in the soil and by soil properties. Additionally, Ni content in plant tissue greatly depends on genotypic ability of plants for Ni uptake. Average Ni content in cereal grains, as per data collected from different countries, varies from 0.34 to 14.60 mg kg^{-1}. In the plant-foodstuffs, the lowest values were reported in apples (0.06 mg kg^{-1} of Ni) and the highest ones in cucumber (2 mg kg^{-1} of Ni) (Kabata-Pendias and Mukherjee 2007). Nickel concentrations in dry leaves of accumulating and hyperaccumulating plant species might reach values as high as 1000 to 35000 mg kg^{-1}.

Several investigators have suggested that Ni might be essential to plants. Its role in the urease metabolism and nodulation of leguminous plants and its effect on the nitrification and mineralization of soil-N has earlier been explored (Eskew *et al.* 1983; Barraquio and Knowles 1989). In comparison to Ni essentiality to plants, its toxicity is well known. Restricted plant growth and the injuries caused by

Table 10 Nickel concentration (mg kg^{-1}) in various medicinal and aromatic plants from different countries.

MAP species	Plant part	Range[a]	Mean (±SD)[b]	Source/Country
Terminalia chebula	as a raw material		0.52	Joseph *et al.* 2011/India
Tinospora cordifolia	as a raw material		13.2	Joseph *et al.* 2011/India
Chamomila recutita	Flower		2.50 ± 0.354	Radanović and Ant.-Mladenović 2008/Serbia
Arnica sp.	Flower		35.6 ± 14.61	Radanović and Ant.-Mladenović 2008/Serbia
Calendulla officinalis	Flower	2.1 – 6.0		Radanović *et al.* 2000/Serbia
Tanacetum parthenium	Flower	0.4 – 2.7	1.62	Marković *et al.* 2008/Serbia
Anethum graveolens	Fruit		3.75 ± 0.75	Radanović and Ant.-Mladenović 2008/Serbia
Vaccinium myrtilus	Fruit[c]	1.02 – 51.2	5.26	Antić-Mladenović *et al.* 2009/Serbia
Rubus ideaus	Fruit[c]	0.05 – 10.0	5.27	Antić-Mladenović *et al.* 2009/Serbia
Hypericum perforatum	Herb[d]	1.0 – 8.0		Radanović *et al.* 2002/Serbia
Ocimum basilicum	Herb	0.9 – 4.1[e]		Zheljazkov and Warman 2003/Canada
Origanum vulgare	Herb		33.3 ± 12.98	Radanović and Ant.-Mladenović 2008/Serbia
Polygonatum verticillatum	Herb	0.54–2.4		Saeed *et al.* 2010
Euphorbia helioscopia L.	Herb		28.6	Hussain *et al.* 2011/Pakistan
Ranunculus mariculatus	Herb		18.2	Hussain *et al.* 2011/Pakistan
Urtica dioica	Herb		15.8	Hussain *et al.* 2011/Pakistan
Achilea millefolium	Herb	4.5 – 14.5		Radanović *et al.*, 2002/Serbia
Datura stramonium	Leaves		4.95 ± 0.23	Olowoyo *et al.* 2012/South Africa
Amaranthus spinosus	Leaves		5.69 ± 0.15	Olowoyo *et al.* 2012/South Africa
Mentha piperita	Leaves	1.7 – 19.2		Radanović *et al.* 2001/Serbia
Salvia officinalis	Leaves	0.6 – 7.6	2.0	Maksimović *et al.* 1999/Serbia
Plantago lanceolata	Leaves		9.70 ± 2.037	Radanović and Ant.-Mladenović 2008/Serbia
Cymbopogon citratus	Leaves	5.37 – 6.23		Haider *et al.* 2004/India
Tanacetum parthenium	Leaves	1.2 – 4.0	2.78	Marković *et al.* 2008/Serbia
Ocimum basilicum	Leaves		36.05 ± 0.01	Annan *et al.* 2010/Ghana
Gymnema sylvestre	Leaves		25.0 ± 0.0012	Annan *et al.* 2010/Ghana
Valeriana officinalis	Root	11 – 48		Radanović and Ant.-Mladenović 2008/Serbia
Althea officinalis	Root	2.2 – 6.0	4.1 ± 1.36	Radanović and Ant.-Mladenović 2008/Serbia
Coix lachryma jobi	Root	6.34 – 12.2		Haider *et al.* 2004/India
Gentiana lutea	Root	27.0 – 58.0	54.0 ± 12.3	Radanović *et al.* 2007/Serbia
Calotropis procera	Root	0.5 – 1.0[f]		Barthwal *et al.* 2008/India
Datura stramonium	Root		10.32 ± 0.09	Olowoyo *et al.* 2012/South Africa
Amaranthus spinosus	Root		15.78 ± 0.14	Olowoyo *et al.* 2012/South Africa
Linum usitatissimum	Seed	0.4 – 6.5		Radanović *et al.* 2000/Serbia
Cardiospermum halicac	Seed	5.46 – 9.64		Haider *et al.* 2004/India
Abutilon indicum	Seed	0.4 – 1.7[f]		Barthwal *et al.* 2008/India
Cyperus rotundus	Tuber	3.74 – 15.03		Haider *et al.* 2004/India
Euphorbia hirta	Whole plant	0.45 – 1.1[f]		Barthwal *et al.* 2008/India

[a] – range as given in related source
[b] - mean (± standard deviation (SD)) as given in related source
[c] – on dry weight
[d]- the term "herb" refers to dry above-ground part of plants used for medicinal purposes
[e] - in container experiment with high Cu compost application
[f]– plants grown at environmentally different sites

the excessive amounts of Ni have been detected (Seregin and Kozhevnikova 2006). Phytotoxic Ni concentrations vary widely among species and cultivars and have been reported to range from 10 to 1000 mg kg^{-1} Ni (Kabata-Pendias and Mukherjee 2007). Furthermore, some plants are very sensitive to increased levels of Ni in the growth media. Even a small concentration of Ni in the nutrient solution, e.g. 1 mg L^{-1} Ni, was reported to be toxic for the hybrid poplar (Punshon and Adriano 1999). On the other hand, many plants, belonging to *Brassicaceae*, *Boraginaceaea*, *Myrtacea*, *Leguminosae* and *Caryophyllacea*, are known for their great tolerance and hyperaccumulation of Ni (Reeves *et al.* 1999; Reeves 2006). Similar to other HMs, Ni in the MAPs varies in respect with characteristics of growing sites and plant genetic potential (**Table 10**).

Radanović *et al.* (2001) revealed a high correlation between soil pH and leaf-Ni content in peppermint, confirming a higher Ni uptake from the acid soils than from the neutral ones. They also reported a strong influence of Ni source on Ni content in the herb. As a consequence of strong aeropollution, Ni in the peppermint herb exceeded 10 mg kg^{-1} in the vicinity of a ferro-nickel smelter (at 1 km distance). These concentrations were 2-3 fold higher compared to those recorded in the herb collected from the locations where soil Ni dominantly originated from geochemical sources. High content of Ni and that of other trace elements (Cd, Co, Cu, Fe and Zn) were reported in marasi (*Curculigo latifolia*) grown at soil contaminated by Cu smelting plant (Mohamed *et al.* 2006).

Soil pollution from geochemical sources might also lead to elevated concentrations of trace elements in the MAPs. Radanović *et al.* (2007) reported high Ni concentrations (24 to 58 mg kg^{-1}) in the gentian roots collected from acid soil developed under the strong influence of serpentine minerals. The galenic forms, prepared from the roots by ethanol extraction, were also rich in Ni (spissum - 40.2 µg g^{-1} Ni; siccum - 45.8 µg g^{-1} Ni). The authors concluded that consumption of gentian galenic forms might cause elevated human intake of Ni, if prepared from the raw material rich in Ni.

Haider *et al.* (2004) reported that Ni content might vary among different MAPs grown under the same geoclimatic conditions; it can also vary among the same species grown under different geoclimatic conditions. In the case of several medicinal plants (*Abutilon indicum*, *Calotropis procera*, *Euphorbia hirta*, *Peristrophe bycaliculata*, *Tinospora cordifoli*), Barthwal *et al.* (2008) found that the level of Ni content might differ in the same plant collected from environmentally different sites of the same city. They concluded that every medicinal plant sample should be tested for contaminant load before processing it further for the preparation of pharmaceutical drugs.

CONCLUSION

In this review, we have provided information on the concentrations of trace elements (heavy metals) in the MAPs as per the reports published from different countries worldwide. A large difference in metal uptake between plant species has been demonstrated in many studies. The growing sites characteristics (particularly soil pH and parent material) and aeropollution are identified as major factors affecting the contents of trace elements in the MAPs and the associated risks of the MAPs contamination. Inherent genotypic characteristics of the species and genotypes of the species also contribute to metals uptake. The risk of human intoxication with trace elements, after the consumption of pharmaceutical drugs (essential oils, extracts, and teas) prepared from polluted raw materials, is generally found to be low.

In order to ensure a good quality of the MAPs and that of the final products (pharmaceutical drugs) so that they are free from potentially harmful constituents to human health, it is necessary to:

- Monitor metallic micronutrients and HMs continuously in soils and the MAPs.
- Choose growing site carefully.
- Manage soil and crops appropriately.
- Choose suitable plant genotypes.
- Define strongly the principles of good agricultural practice (GAP in MAPs) and/or
- Grow the MAPs according to the principles of organic farming.

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Appendix List of the medicinal and aromatic plants presented in this review

Latin name	Common name
Abutilon indicum	Indian Mallow
Achilea millefolium	Yarrow
Alchornea cordifolia	Christmas tree
Althea officinalis	Marshmallow
Alyssum bertoloni	-*
Amaranthus spinosus	Spiny amaranth, Prickly amaranth, Thorny amaranth
Ananas comosus	Pineapple
Anethum graveolens	Dill
Angelica arhangelica	Angelica
Arnica Montana	Mountain arnica
Artemisia absinthium	Absinthium, Absinthe wormwood
Artemisia dracunculus	Tarragon
Asparagus somnifera	-
Azadirachta indica	Neem
Bacopa monnieri	Brahmi, Water hyssop
Betula sp.	Birch
Bidens tripartita	Bur-marigold
Borago officinalis	Borage
Boswellia serrata	Indian Olibanum Tree, Olibanum, Luban, Gond
Calendulla officinalis	Marigold
Calotropis procera	Calotrope
Camellia sinensis	Black tea
Cardiospermum halicacabum	Balloon plant or Love in a puff
Centella asiatica	Commonly centella
Chamomila recutita	Camomile
Chenopodium foliosum	Goosefoot, Pig Weed
Cochicum luteum	-
Coelus forskohlii	False Boldo, Coleus, Forskohlii, Plectranthus
Coix lachryma jobi	Job's Tears, Coixseed
Coriandrum sativum	Coriander
Crategus sp.	Hawthorn
Curculigo latifolia	Marasi
Curcuma longa	Turmeric
Cymbopogon citratus	Lemongrass
Cyperus rotunuds	Mustaka, Red Nutsedge, Coco Grass
Datura stramonium	Jimson weed, Devil's trumpet
Desmodium adscendes	Amor seco, Amor-do-campo
Echinacea purpurea	Echinacea
Euphorbia helioscopia L.	Sun Spurge
Euphorbia hirta	Cats hair, Asthma weed
Fagopyrum esculentum	Buckwheat
Foeniculum vulgare	Fennel
Gentiana lutea	Yellow gentian
Gingko biloba	Ginkgo
Glycyrrhiza glabra	Liquorice
Gymnema sylvestre	Gymnema
Helianthus annuus	Sunflower
Hypericum hirsutum	Hairy St John's Wort
Hypericum perforatum	St. John's Wort
Hyssopus officinalis	Hyssop
Lavandula angustifolia	Lavender
Leonurus cardiaca	Motherwort
Levisticum officinalis	Lovage
Linum usitatissimum	Linseed
Lippia multiflora	Savannah tea
Malva silvestris	Mallow leaves
Marrubium vulgare	Horehound
Melissa officinalis	Lemon balm
Mentha arvensis	Japanese mint
Mentha piperita	Peppermint
Nasturtium officinale	Watercress
Ocimum basilicum	Sweet basil
Orig. vulgare spp.hirtum	Italian Oregano, Greek Oregano
Origanum heracleoticum	Greek Oregano
Origanum onites	Cretan Oregano, Turkish Oregano

* to our knowledge, no common name in English